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Tracer transport in the tropical stratosphere due to vertical diffusion and horizontal mixing

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Abstract.
Observations of linearly increasing (e.g., SF_6) and periodically varying (e.g., H_2O+2CH_4) long-lived tracers in the lower stratosphere provide independent constraints on theories of transport in the region. Taken together, these data allow separation of the roles of diffusion (with coefficient K_θ) and advection (at rate Q) through isentropic surfaces, and mixing of extra-tropical air into the tropics (with relaxation time τ). Using a one-dimensional diffusive-advective model of the tropical stratosphere, which allows relaxation of mixing ratios to extra-tropical values, we obtain solutions for periodic and linear tracers. Fitting the solutions to observations yields $K_\theta \approx 0.3$ K^2/day ($K_z \approx 0.01$ m^2/s), $\tau \approx 1.3$ years, and $Q \approx 0.5$ K/day ($\overline{w} \approx 0.3$ mm/s). These values produce profiles for CO_2 in reasonable agreement with aircraft observations. However, a large range of K_θ results in equally good agreement, although τ and Q are more tightly constrained. In the lower tropical stratosphere, vertical diffusion appears to play little role in transporting tracer.

1. Introduction

The rate at which lower stratospheric extra-tropical air mixes into the tropics and is lofted upwards determines the extent to which midlatitude aircraft emissions will affect the ozone layer. Although aerosol observations [*Trepte and Hitchman*, 1992] suggested a mixing barrier between the tropics and extratropics, an idea formalized by *Plumb* [1996], photochemical calculations show the tropics cannot be completely isolated [*Avallone and Prather*, 1996]. Recently, several studies have estimated rates of mixing and ascent using aircraft [*Boering et al.*, 1996; *Minschwaner et al.*, 1996; *Volk et al.*, 1996] and satellite measurements [*Mote et al.*, 1996; *Schoeberl et al.*, 1997; *Randel et al.*, 1997].

One way to estimate transport parameters is to observe the vertically propagating signal in the tropics of conserved tracers whose mixing ratio varies periodically (e.g., $\hat{H} = H_2O+2CH_4$ and CO_2). However, the oscillation amplitude alone cannot pin down a horizontal mixing timescale, as it is attenuated by both the presence of extratropical air and by vertical diffusion. Limiting bounds on the mixing rate and diffusion have been estimated from the $\hat{H}$ amplitude; e.g., *Mote et al.* [1996] assumed no mixing to obtain an upper bound on the rate of diffusion, while *Randel et al.* [1997] assumed no diffusion to obtain an upper bound on the rate of mixing. Recently, Phil Mote (personal communications; 1997) has used H_2O and CH_4 separately to estimate rates of diffusion and mixing simultaneously.

Hall and Waugh [1997] showed that the phase lag of a periodic signal and the mean age (the stratospheric lag time to a tracer varying linearly in the troposphere [e.g., *Hall and Plumb*, 1994]) are timescales characterizing different properties of transport. In this paper we show how these two timescales supplement the amplitude information, thus providing three independent constraints on the roles of horizontal mixing, vertical diffusion, and vertical advection in the tropics. We estimate these parameters using analyses of $\hat{H}$ from the Halogen Occultation Experiment (HALOE) instrument aboard Upper Atmosphere Research Satellite (UARS) [*Randel et al.*, 1997], and of SF_6 by the ACATS instrument [*Elkins et al.*, 1996] during the Airborne Southern Hemisphere Ozone Experiment (ASHOE) and Stratospheric Tracers of Atmospheric Transport (STRAT) campaigns.

2. One Dimensional Model

In order to extract rates of diffusion, mixing, and vertical advection from tracer observations, we use the simplest model possible that includes these effects. We solve the one-dimensional tracer continuity equation

$$\frac{\partial \chi}{\partial t} + Q\frac{\partial \chi}{\partial \theta} - \frac{1}{\rho}K_\theta\frac{\partial}{\partial \theta}(\rho\frac{\partial \chi}{\partial \theta}) = -\frac{1}{\tau}(\chi - \chi_e) \quad (1)$$

where χ is a tracer mixing ratio, K_θ a coefficient of vertical diffusion, Q a vertical velocity, τ a relaxation time to the specified extra-tropical mixing ratio χ_e, summarizing the mixing rate of air into the tropics, and ρ the air density falling exponentially with scale height H_θ. The vertical coordinate is potential temperature θ above the tropical tropopause (assumed to be at 390K). In the atmosphere, K_θ, Q, and τ vary with space and time, but here we consider only constant values.

We solve (1) for two boundary conditions: (i), a sinusoidal source ($\chi(0,t) = \cos\omega t$), and (ii), a tracer of

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linearly increasing abundance ($\chi(0,t) = ct$). The periodic solution has amplitude A and phase lag τ_ω, while the linear solution determines the mean age Γ. In each case, the extra-tropical value χ_e must be specified. For the periodic tracer, we take $\chi_e = 0$, as observations of $\hat{H}$ show small amplitude in the extra-tropical stratosphere compared to the tropics [*Randel et al.*, 1997]. For the linear tracer, we take $\chi_e = c(t - \Gamma_e(\theta))$, where $\Gamma_e(\theta) = m\theta + \Gamma_e(0)$ is an extra-tropical vertical age profile with a constant vertical age gradient m.

The explicit solutions are shown in Table 1. For low diffusion ($\tau_\theta\omega << 1$, $Q >> K_\theta/H$, and $\tau >> \tau_\theta$) the amplitude $A \sim e^{-\theta/\tau Q}$ and the phase lag $\tau_\omega \sim \theta/Q$. In this limit, diffusion plays a negligible role in propagating the annual cycle compared to advection, and also a negligible role in attenuating the cycle compared to relaxation to the extra-tropical value.

Table 1. Explicit Solutions

periodic boundary condition $\chi(0,t) = \cos\omega t$:
$\chi(\theta,t) = A\cos\omega(t - \tau_\omega)$
where $A = e^{-k_A\theta}$, $\tau_\omega = k_\omega\theta$,
$k_A = (\tilde{Q}/2K_\theta)\left(\psi\cos\left(\frac{1}{2}\tan^{-1}\gamma\right) - 1\right)$,
$k_\omega = (\tilde{Q}/2K_\theta\omega)\psi\sin\left(\frac{1}{2}\tan^{-1}\gamma\right)$,
$\psi = (\tau_\theta\omega)^{1/2}(1+\gamma^{-2})^{1/4}$,
$\gamma = \omega/(\tau_\theta^{-1} + \tau^{-1})$, $\tau_\theta = 4K_\theta/\tilde{Q}^2$, and
$\tilde{Q} = Q + K_\theta/H_\theta$

linear boundary condition $\chi(0,t) = ct$:
$\chi(\theta,t) = c(t - \Gamma)$
where $\Gamma = (\Gamma_e(\theta) - \delta\Gamma) - (\Gamma_e(0) - \delta\Gamma)e^{-\theta/L}$,
$\Gamma_e(\theta) = m\theta + \Gamma_e(0)$, $\delta\Gamma = \tau(m\tilde{Q} - 1)$, and
$L = (2K_\theta/\tilde{Q})\left(\sqrt{1+\tau_\theta/\tau} - 1\right)^{-1}$

3. Observations

The HALOE instrument measures stratospheric H_2O and CH_4, and the quantity $\hat{H}$ is well conserved in the lower and middle stratosphere [*Dessler et al.*, 1994]. $\hat{H}$ varies seasonally at the tropical tropopause, and this signal is transported vertically through the tropical stratosphere [*Mote et al.*, 1995; 1996]. *Randel et al.* [1997] have fit annual and semi-annual harmonics to the HALOE $\hat{H}$ time series from 1991 to 1996. We use their phase and peak-to-peak amplitude at 100 hPa to 31 hPa to obtain $k_A = 0.0043$ K^{-1} and $k_\omega = 0.0050$ yr/K for the coefficients defined in Table 1, giving a 55% attenuation and an 11 month phase lag at 31 hPa.

SF_6 is nearly inert and its abundance is increasing approximately linearly [*Maiss et al.*, 1996; *Geller et al.*, 1997]. Here we use SF_6-inferred age from ASHOE and STRAT data to estimate mean age [*Elkins et al.*, 1996; *Waugh et al.*, 1997; *Volk et al.*, 1997]. Using the surface mean time series from *Geller et al.* [1997] and a tropopause age of 0.8 year [*Volk et al.*, 1997], we obtain, in the tropics (±15°) at 80K (±5K) above the tropopause, $\Gamma = 0.9 \pm 0.3$ years with respect to the tropopause. To obtain an estimate of $\Gamma_e(0)$, we average mean ages between 0 and 20K from the height of the tropical tropopause (i.e., 390 and 410K) at latitudes between 20° and 40°, finding $\Gamma_e(0) = 0.9 \pm 0.4$ years, again with respect to the tropical tropopause. Linear fits of the vertical age variation in the extra-tropics produce $m = 0.025$ yr/K. Finally, $H = 7$ km is converted to $H_\theta = 162$ K for use in (1) by employing the close linear relation between $\log p$ and θ of ASHOE and STRAT data in the tropical lower stratosphere.

4. Solving for K_θ, τ, and Q

A and τ_ω (Table 1) may be used to eliminate Q, thereby providing a relation between K_θ and τ. This "A-τ_ω curve" is drawn as a heavy solid line in Figure 1. There are two limits to note. For $\tau \to \infty$ (complete isolation of the tropics), vertical diffusion is solely responsible for the observed attenuation of the periodic tracer signal. This upper bound on K_θ is about 1.8 K^2/s ($K_z \approx 0.04$ m^2/s), within the range obtained by *Mote et al.* [1996]. In the opposite limit, $K_\theta \to 0$, all the attenuation of the periodic signal is due to mixing of extra-tropical air into the tropics. τ in this limit is about 1.2 years, a lower bound consistent with *Randel et al.* [1997].

A second relation between K_θ and τ is obtained by eliminating Q from τ_ω and Γ (Table 1), producing a "Γ-τ_ω curve". This curve is shown in Figure 1 as a heavy dashed line. The A-τ_ω and Γ-τ_ω curves intersect at $\tau \approx 1.3$ years and $K_\theta \approx 0.3$ K^2/day ($K_z \approx 0.01$ m^2/s). The simultaneous solution for the vertical velocity is $Q \approx 0.5$ K/s ($\overline{w} \approx 0.3$ mm/s). This represents our best estimate for these values.

The best estimate values are in the low diffusion limit. The phase velocity θ/τ_ω of the HALOE $\hat{H}$ signal only differs from Q by 1%, the first order correction to Q being $4K_\theta/(Q\tau)$, which is 100 times smaller. The effect of diffusion on the amplitude is greater, the first order correction to $k_A \sim (\tau Q)^{-1}$ being $\omega^2 K_\theta/Q^3$, which is about 10 times smaller: for best estimate values, $A(80\text{K}) = 0.70$, while for the same τ but $K_\theta = 0$, $A(80\text{K}) = 0.73$.

The circles of Figure 1 are solutions obtained by varying A, τ_ω and Γ by ±10% about the fits to observations. Some combinations (not plotted) shift the Γ-τ_ω and A-τ_ω curves far enough apart that there is no intersection. Q is insensitive to A and Γ, but varies nearly inversely with τ_ω. Despite these uncertainties, it is encouraging that $\tau = 1.3$ years is within the range 1.1 ± 0.2 years obtained by *Volk et al.* [1996] in an independent analysis, and $\overline{w} \approx 0.3$ mm/s falls within the uncertainty range of the annual mean vertical velocity in the tropics estimated by *Rosenlof* [1995].

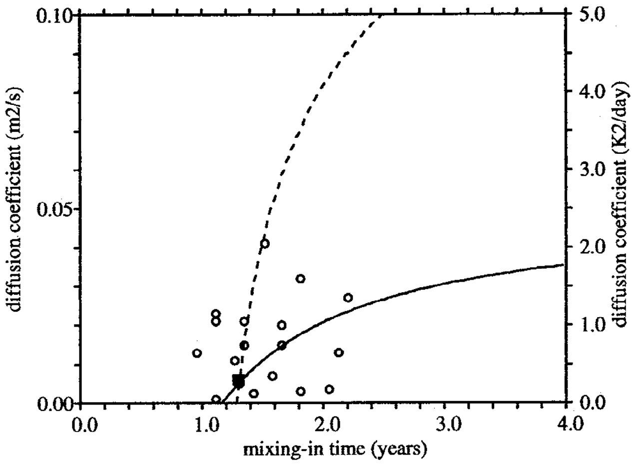

Figure 1. Curves of the vertical diffusion coefficient K_θ versus the mixing-in time τ derived from fits to satellite and aircraft observations. Solid and dashed lines represent independent techniques. The best estimate solution is indicated at the intersection point by the dark square symbol. Circles are solutions using 10% variations of observations about the fits. See text for details.

5. Predicted CO_2 Profiles

Measurements of stratospheric CO_2 [*Boering et al.*, 1994; 1996] are independent data testing our estimates of K_θ, τ, and Q. CO_2 exhibits stratospheric gradients due to both a long term quasi-linear trend and an annual cycle in the troposphere. Thus, we combine solutions for oscillating and linear tracers to obtain a modeled CO_2 profile. As a boundary condition, we fit a linear trend and an annual and semi-annual cycle to the observed tropopause time series of *Boering et al.* [1996].

Figure 2 shows the aircraft CO_2 measurements on Febuary 1996 and October 1994 over the first 100K of potential temperature from the tropical tropopause within 15° of the equator. The solid line is the model solution for the best estimate values from the HALOE and SF_6 data. There is good agreement in the position of the minimum and maximum. The October model profile is a remarkable fit to the measurements, while the February profile is similar in shape to the measurements but shifted by about 1 ppm. There are fewer tropical tropopause measurements over the year preceding February 1996 than during that preceding October 1994, so that the boundary condition is less accurately determined. In general, the model profiles are quite sensitive to the tropopause boundary condition. The observed February profile also show a sharp fall-off above 80K. These data were taken further from the equator than the others, and may represent a recent extra-tropical intrusion.

The minima and maxima of the model profiles ascend at different rates, even though there is no variation in the phase velocity, the result of adding an attenuating sinusoid to a non-constant background [*Waugh et al.*, 1997]. Agreement in the height of profile extrema in seasons of opposite phase, despite the constant coefficients of the model, suggests that observed variations in the ascent rate of the CO_2 cycle [*Boering et al.*, 1996] may not be due entirely to seasonal variations in vertical velocity. For our best estimate values, the February minimum propagates upward from the tropopause about 25% faster than does the October maximum (compared to a 60% difference observed by *Boering et al.* [1996]). Reducing τ to 1.0 year results in a 40% difference. Because the minimum versus maximum effect is of comparable magnitude to seasonal $\overline{w}$ variations, it should be accounted for, if possible, when inferring $\overline{w}$ from CO_2 observations. In principle, this can be accomplished by removing the CO_2 variation due to the Γ fall-off before tracking the cycle extrema.

To test sensitivities, we computed model CO_2 profiles for other values of K_θ, τ, and Q (Figure 2). Doubling τ worsens the agreement with observations, as does a 20% reduction in Q. On the other hand, tripling K_θ has very little effect. This is consistent with the low diffusion limit. Vertical diffusion plays little role in tracer transport, and hence tracer observations provide little information about the rate of diffusion.

6. Conclusions

Using satellite $\hat{H}$ measurements, aircraft SF_6 measurements, and a simple diffusion-advective model of the lower tropical stratosphere that allows relaxation to the extra-tropics, we have separated the roles of vertical diffusion and extra-tropical mixing on trace gas signals

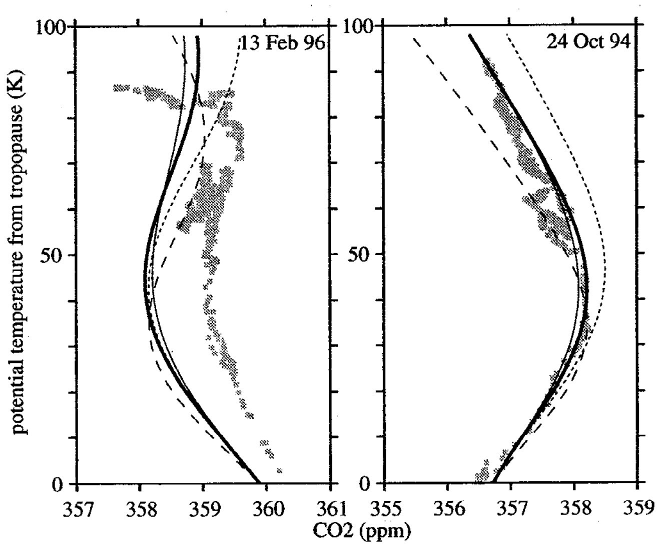

Figure 2. Tropical profiles of CO_2 for February 1996 (left panel) and October 1994 (right panel). The symbols are measurements from aircraft. The heavy solid line is the model using our best estimate parameter values. Other curves use best estimates, but with K_θ tripled (light solid), τ doubled (dotted), and Q decreased 20% (dashed).

in the tropics. Our best estimate from this anaysis is $K_\theta \approx 0.3$ K^2/day ($K_z \approx 0.01$ m^2/s), $\tau \approx 1.3$ years, and $Q \approx 0.5$ K/day ($\overline{w} \approx 0.3$ mm/s). The uncertainty in the measurements, and the limits of such a simple model, imply a large range of equally possible values (at least a factor 3 for K_θ) about this best estimate. However, the estimates of τ and $\overline{w}$ agree well with independent estimates of *Volk et al.* [1996] and *Rosenlof* [1995], respectively. In addition, these values of K_θ, τ, and Q produce model CO_2 profiles in good agreement with CO_2 measurements in the lower stratosphere in different seasons, despite the fact that the model employs constant coefficients.

Our analysis has implications for two- and three-dimensional models of the stratosphere. It is a great challenge for such models simultaneously to simulate accurate values of Γ, A, and τ_ω in the tropics. *Hall and Waugh* [1997] show two GCMs with greatly different Γ which both significantly overattenuate the annual cycle amplitude. Preliminary analysis similar to that presented here, but applied to model data, suggests that a major problem is too much vertical diffusion in the tropics, perhaps by an order of magnitude. However, once models are in a low diffusion regime, the precise value for K_θ is not critical. The condition for acheiving this regime may be estimated from the first order correction to the low diffusion limit for the amplitude, and is $K_\theta << Q^2/\omega^2\tau$, or, for our best estimate values, $K_\theta << 5$ K^2/day ($K_z << 0.1$ m^2/s).

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